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POTENTIAL METHODS FOR METHANE EXTRACTION FROM GEOPRESSURED BRINE AT HIGH TEMPERATURE AND PRESSURE

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ABSTRACT

Recovery of methane from Gulf Coast geopressured-geothermal reservoirs does not appear to be profitable without a rise in natural gas prices to offset high production costs. Fluid disposal by injection even into shallow aquifers is also expected to be costly. If injection into the production reservoir becomes necessary to maintain productivity and to minimize subsidence, the injection pumping costs approach and even exceed the value of the recoverable methane. An option aimed at reducing the injection pump operating costs is to maintain a higher than normal pressure at the production wellhead to reduce the injection-pumping work load. This option, however, is considerably less attractive if that portion of methane still dissolved at elevated pressure cannot be recovered. Therefore, there is a strong incentive to devise methods for extracting methane at high pressures and temperatures. Several methods have been identified and examined for technical feasibility, potential benefits, and problems of implementation. Liquid extraction with a very low water-soluble organic is a technically feasible method and looks promising as an applicable process. A candidate solvent is hexadecane, a paraffinic hydrocarbon with the necessary phase-equilibrium thermodynamic properties to satisfy the technical requirements for such an operation, without any obvious economic barriers. Gas stripping is another technically feasible method, but the economics do not look favorable because of gas dissolution losses. Freon refrigerants were considered because of their ease of product-stripping gas separation and nitrogen was considered because of its low cost. Brine-driven positive displacement pumps and hydraulic turbines directly coupled to pumps with provisions for methane exsolution are technically feasible concepts and could eliminate or greatly reduce pump power costs. These extraction operations will not preclude the option of recovering the thermal energy component, if desired. More detailed analyses of these methods including measurements of mutual solubilities, tests of small extraction columns, design and tests of prototype machines

References and illustrations at end of paper.

and gas separators, and estimates of process costs are now in progress.

INTRODUCTION

Recent technical and economic assessments of methane (CH₄) production from geopressured-geothermal reservoirs in the Texas and Louisiana Gulf Coast area have been made by Swanson and Osoba, and Doscher et al. They conclude that to obtain a reasonable return on capital for CH₄ recovery from reservoirs with properties shown in Table 1, the CH₄ selling price must be in the range of \$7.50 to \$9.24/Mcf.

Swanson and Osobal further state that the \$7.50/Mcf will produce only a small profit and is based on optimistic reservoir parameters. Doscher et al.² emphasize that a great deal of optimism was used to arrive at their \$9 to \$10/Mcf selling price. Samuels3 also concludes that "unless the methane content and market value are sufficient to offset the cost of the production and reinjection wells, there is currently little incentive to develop this resource." It is clear that at the present value of methane (~\$3.50/Mcf), commercialization of the geopressured-geothermal resource will not be expected in the near future. Exploitation of the resource will indeed be unlikely, barring unforeseen circumstances such as the discovery of large quantities of free CH, gas or highly productive reservoirs, unless the CH4 selling price rises steeply relative to expenses or significant reductions in production costs are achieved. Production and injection well capital costs comprise the major share of expenses, but the operating and maintenance (O&M) costs for injection are also significant. Doscher et al. approximate the O&M costs for injection with the following formula:

$$\frac{5}{bb1} = \frac{0.02 + 0.005(\frac{mcf}{P})}{0.005(\frac{mcf}{P})}$$

where \$/Mcf is the selling price of CH₄, and P is the required injection pressure in 10³ psi. Pump power costs are proportional to injection pressure and the value of CH₄. The \$0.02 constant accounts for pump maintenance costs.

At 40 scf CH₄/bbl brine, the value of CH₄/bbl brine is (0.04)(\$/Mcf). Assuming that the pressure required for injection into shallow aquifers (6000-ft deep) is 1000 psi, then the ratio of injection O&M costs to the value of CH₄ is given by

$$R = 0.5/(\$/Mcf) + 0.125 \dots (2)$$

The injection O&M costs relative to the value of CH₄ are, from equation (2), between 17.5% for \$10/Mcf gas and 26.8% for \$3.50/Mcf gas, clearly a significant percentage. The O&M costs are predominately pump power and maintenance costs, which rise steeply with required injection pressure and become prohibitively high for geopressured waters with low CH₄ content.

It is clear that measures that could reduce injection costs would contribute significantly to the economic viability of geopressured resource development. A potential option in the production of geopressured aguifers is to maintain sufficient pressure at the wellhead to reduce the injection pump workload. For shallow injection horizons, direct injection without pumping may be feasible. If it becomes necessary to inject into the production reservoir for pressure maintenance and subsidence control, injection pumping O&M costs could easily exceed the value of recoverable CH4. Again, by maintaining higher than normal production wellhead pressures, the power requirements for injection pumping could be substantially reduced to a level where an economic advantage is gained even though production rates are also reduced. However, the potential savings would be offset by the loss of CH4 still dissolved at the elevated pressures. There is a large incentive, therefore, to be able to extract CH4 at high pressures and also at production fluid temperatures to permit recovery of the thermal energy component from the CH4-depleted brine.

There are several potential methods for extracting dissolved CH4 at produced-brine temperatures of 150°C and at anticipated pressures of 1000 to 1500 psi for injection into 6000-ft deep aquifers. The chemical techniques include gas stripping, which is technically viable but may not be economical, and solvent extraction. Mechanical methods consist of positive-displacement hydraulic motors and hydraulic turbines coupled directly to pumps with provisions for CH4 exsolution. Although the ensuing discussion addresses CH4 extraction methods at pressures required for injection into shallow aquifers, these methods, in theory, should be applicable at the higher pressures necessary for injection into the production reservoir and may indeed be of greater value.

GAS STRIPPING

Gas stripping or desorption is a common chemical process for recovery of dissolved gases in liquid streams. It should be possible to strip dissolved CH₄ from geopressured waters by application of this technique. In order to maintain pressure for direct injection purposes, desorption of CH₄ would be accomplished at injection pressures between 1000 and 1500 psi. The stripped CH₄ is recovered after separation from the working gas, while the spent liquid (still under pressure) is disposed of by injection without the necessity for pumping. The economic viability of this pro-

cess depends on identifying a stripping gas that is either exceedingly inexpensive or has minimal solubility in geopressured brines.

Obviously, air would be the least expensive gas stripping agent. However, there are at least two critical limitations. First, the formation of an explosive air-methane gas mixture would not be acceptable. Second, the corrosiveness of the brine would greatly increase with aeration.

It should be emphasized that CH₄ solubility, even in pure water, is only 10 scf/bbl at 150°C and 100 atm.^{5,6} In 16 wt% sodium chloride (NaCl), CH₄ solubility at the same temperature and pressure is significantly reduced to about 5 scf/bbl.⁶ The lower CH₄ concentration would significantly raise the necessary profitable CH₄ selling price that was discussed above. At 10 scf/bbl, CH₄ is worth \$0.035/bbl (at \$3.50/Mcf). The value of any stripping agent that is dissolved and presumably lost in the spent brine must therefore be substantially less than \$0.035/bbl brine.

Another way to view the economics is to compare the value of CH $_{\rm h}$ to bulk liquid nitrogen (liquid N $_2$). The value of CH $_{\rm h}$ at \$3.50/Mcf translates to \$0.079/lb, while liquid N $_2$ delivered in quantities greater than 1 MMcf/mo costs about \$0.04/lb. In short, because CH $_{\rm h}$ is so relatively inexpensive and its equilibrium concentration in the brine so low, the choice of a stripping agent is critical and severely limited. Another consideration is the cost of the subsequent gas separation process that is necessary to recover pipeline quality CH $_{\rm h}$, as well as the stripping agent for recycle. We have considered separation methods ranging from selective CH $_{\rm h}$ absorption to condensation of either CH $_{\rm h}$ or the stripping agent.

Halogenated Hydrocarbons

The first stripping agents we considered were the halogenated hydrocarbons, namely Freon* refrigerants. Freon would be completely vaporized on contact with the hot brine, producing excellent gas-liquid interfacial contact for stripping dissolved CH4. Gaseous Freon is condensed and separated from CH4 at a temperature where the partial pressure of Freon in the recovered CH4 stream is acceptably low in terms of cost and CH4 purity.

However, only a few Freon compounds have the desired thermodynamic and thermochemical properties to suffice as potential stripping agents. The stripping agents must be sparingly soluble in the geopressurized brines at temperatures of $150\,^{\circ}\text{C}$ and at pressures of about 100 atm. They must be thermally and chemically stable in the brine environment. Finally, their critical temperatures must be less than $150\,^{\circ}\text{C}$ to assure complete vaporization for stripping purposes, yet have low volatility for ease of condensation in the gas separation and purification step.

The difficulty in selecting an appropriate stripping agent is partly because of the scarcity

*Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Department of Energy to the exclusion of others that may be suitable. of solubility data in the literature at these temperatures and pressures. In our initial search, we found that dichlorotetrafluoroethane (Freon 114) met some of the criteria. Its critical temperature is 145.7°C and its normal boiling point is 3.77°C.8 However, at 100 atm and 150°C, its solubility in brine may be too high to be cost effective. Stepakoff and Modica measured the solubility of Freon 114 and other halocarbon refrigerants in aqueous systems containing as much as 7 wt% NaCl between 0 and 50°C. From the correlation given by Prausnitz, 10 we plotted the Freon 114 solubility data versus 1/temp on semilog coordinates in Fig. 1 and extrapolated to 150°C to give a solubility of about 4.4 ppm/atm. Assuming a Henry's law relationship is valid to 100 atm, then the calculated solubility of Freon 114 at that pressure in 7 wt% NaCl at 150°C is about 440 ppm. At \$1.80/1b for Freon 114 (see Ref. 11), the value of Freon 114 dissolved in the brine and presumably not recoverable exceeds the value of CH4 by eightfold.

Other halogenated hydrocarbons are not likely to be better. Partially halogenated hydrocarbons are considerably more soluble than completely halogenated hydrocarbons, $^8,^9,^{12}$ so it appears that at a partial pressure of 100 atm only the cheapest gases such as N_2 would even begin to approach economic feasibility.

Determination of relative process flow rates and number of stages in an ideal countercurrent operation can be made, however, for stripping by any gas. An extrapolation of O'Sullivan's and Smith's data on CH4, solubility in 0 to 4M NaCl at 51.5 to 125°C and 100 to 600 atm gives an average Henry's constant of 1.47×10^5 atm for CH₄ in 3M NaCl at 150°C and 0 to 100 atm. Using this value to construct the equilibrium curve, we determined the number of ideal stages for a countercurrent multistage operation. With 0.56 lb Freon 114/ 100 lb brine, which corresponds to twice the minimum gas requirements, between three and four ideal stages are required for 90% recovery of CH4 from brine. The stage efficiency depends on the design characteristics of the stages and the properties and flow rates of the fluids. For absorption or stripping of sparingly soluble gases, tray efficiencies can be as low as 10%. 13 Therefore, a stripping operation may require towers with 30 to 40 stages. In practice, it may be more cost effective to use fewer stages and higher gas-toliquid ratios.

Similarly, it was determined that approximately four transfer units are required in a continuous differential contractor, assuming the worst case where the principal diffusional resistance resides in the liquid phase. The number of transfer units is an indicator of the ease or difficulty of the mass transfer operation.

The gas stripping operation with Freon 114 is illustrated in Fig. 2. The process flows merely illustrate relative quantities based on the equilibria data, 90% CH4 recovery, and twice the minimum gas-liquid ratios that were used above for stage determination. Freon is condensed at a conservatively low temperature to minimize vapor losses in the CH4 recovery stream and then recycled.

Nitrogen

Liquid N_2 delivered by truck in quantities on the order of 10 MMcf/mo costs approximately \$0.04/lb plus transportation. Large users able to absorb the capital costs of a N_2 pipeline can reduce the costs to perhaps \$0.02/lb. Although the cost of liquid N_2 is less than CH₄ selling at \$3.50/Mcf (\$0.079/lb), its solubility at 150°C, extrapolated from data between 51.5 and 125°C, is roughly equal to that of CH₄ on a weight basis. Therefore, at \$0.04/lb, 50% of the recoverable CH₄ value would be lost by dissolution of N_2 gas in the brine. With these inherent losses, stripping CH₄ with N_2 is not promising unless CH₄ prices were to rise dramatically faster than liquid N_2 prices. This is a conceivable, but unlikely event.

In the event that N_2 could be used, then the stripped CH4 could be separated from N_2 by preferential absorption with an appropriate hydrocarbon. Methane in essentially pure form would be recovered by simple depressurization of the hydrocarbon. Both the solubility of N_2 in the hydrocarbon and the vapor pressure of the hydrocarbon must be sufficiently low to minimize loss of N_2 and the hydrocarbon during the CH4 recovery step. Low-vapor-pressure paraffinic hydrocarbons would be excellent absorbants for CH4. However, the solubility of N_2 in, for example hexadecane, is also quite high and may be indicative of the lack of absorbants which are selective for CH4 in mixed gases.

Figure 3 illustrates the gas stripping operation using N_2 gas. The process flow rates are based on the same assumptions as the Freon 114 operation. As indicated, CH₄ could be separated from N_2 by liquefaction or by preferential absorption. However, liquefaction may be inefficient and energy intensive because of the large fraction of noncondensable N_2 gas. Absorption of CH₄ would be feasible, provided an absorbant selective for CH₄ could be identified.

SOLVENT EXTRACTION

The use of a paraffinic hydrocarbon for direct contact with the geopressured brines to extract dissolved CH₄ looks promising. The process, conceptually, is quite simple. A high-boiling, paraffinic hydrocarbon is contacted with the brine in an extraction tower. Methane, being more soluble in the hydrocarbon, is extracted and subsequently recovered in essentially pure form by depressurization of the extract. Solvent loss is controlled by selecting a low-vapor-pressure compound.

A promising hydrocarbon candidate is hexadecane, which has a vapor pressure of 10 mm Hg at 149.8°C and a solubility in water that is presumably quite low. We estimated hexadecane solubility using McAuliffe¹⁵ 25°C solubility data on C₁ to C₈ normal paraffinic hydrocarbons, extending to C₁₆ his interpretation that on an equal hydrocarbon vapor pressure basis, approximately the same weight of paraffinic hydrocarbon dissolves in water. The solubility of hexadecane was, therefore, roughly estimated by multiplying the n-octane solubility at 25°C by the ratio of the vapor pressure of hexa² cane at 150°C to the vapor

pressure of n-octane at 25°C. This gave a value of about 0.5 ppm for hexadecane solubility at 150°C. Using this figure and a recent cost quotation of \$4/lb for 95% pure hexadecane, 16 the value of hexadecane that is dissolved and nonrecoverable in the brine would be an acceptable 4.5% of the value of recovered CHa.

For the process to be feasible, the solubility of CH4 in hexadecane must be significantly higher than in the brine to effect an extraction. Cukor and Prausnitz report Henry's constants for CH4 in hexadecane in the temperature range of 25 to 200°C . Again assuming that Henry's constant is independent of pressure up to 100 atm, a solubility of 52,000 ppm is calculated for CH4 in hexadecane at 100 atm and 150°C. This compares with about 540 ppm for CH4 in 16 wt% NaCl at the same temperature and pressure. Unlike gas stripping, solvent extraction can be more selective in removal of CH4. This would be significant in high carbon dioxide (CO2) brines where removal of CO2 could elevate the pH-initiating precipitation of dissolved minerals. But like N2, CO2 is also readily soluble in hexadecane.

Again using the data of O'Sullivan and Smith for CH4 solubility in 3M brine, and those of Cukor and Prausnitz for CH4 in hexadecane at 150°C, the number of ideal stages is determined for a countercurrent multistage operation for extraction of CH4 from brine. For 90% CH4 recovery and 2.26 lb hexadecane/100 lb brine (corresponding to twice the minimum solvent requirements), between three and four ideal stages would be required to effect the separation. As previously described for the gas stripping operations, stage efficiencies are highly variable. The number of actual stages will depend on stage design, fluid properties, and process variables.

For continuous differential contact operation, we calculated about three or four ideal transfer units would be needed. The number is weakly dependent on the phase, solvent or brine, that controls diffusional mass transfer.

Figure 4 illustrates the liquid extraction operation for the conditions just described. The operation is in principle quite simple. Yet the tower design can be quite complex because of potential problems that may arise from emulsion formation and hence difficulties in phase separation.

Formation of stable emulsions is a major concern in liquid-liquid extraction processes. Small bubbles of expensive solvent rejected with spent brine would be unacceptable. Low concentrations of surfactants may be necessary to assist in coalescing the dispersed phase. In addition, holding tanks may be required to allow settling, coalescing, and optimal phase separation. Upon separation, CH, is recovered by simply releasing the pressure on the extract.

An intriguing idea that could favorably affect the economics of the process is to utilize the almost unlimited depth of the production well as a cocurrent extraction column. The extractant hexadecane would be injected into the well via a small-diameter tubing string. The long contact times and turbulent flow conditions would be favorable for effective mass transport. Phase separation would be accomplished on the surface.

MECHANICAL METHODS FOR METHANE RECOVERY

The simplest technique for recovering dissolved CH₄ is to lower the pressure to ambient and collect the evolved gases. The principal disadvantages, as previously mentioned, are the added pumping costs required for brine injection and the loss of thermal energy if pressure is fully reduced to 1 atm and hence about 100°C. Furthermore, if positive-temperature soluble species are present, they may precipitate, creating fluid handling problems.

There are several potential methods that utilize the hydraulic energy available in the liquid discharged from the separator, which would operate at pressures somewhat below the wellhead but sufficient to drive the fluid-injection operation. The methods to be discussed are variations of positive-displacement hydraulic engines and hydraulic turbines with special provisions to permit CH₄ exsolution and recovery.

Hydraulic-Driven Membrane Pump

The hydraulic-driven membrane pump is a sequential depressurization-repressurization process. A conventional gas separator is used for initial CH, separation from wellhead fluid. Pressure in that separator is adjusted to satisfy injection pressure requirements, pressure losses in surface equipment, and pressure requirements that may be dictated by brine chemistry. Brine from the separator passes alternately through at least two depressurization-separation vessels in parallel. At any moment one of the vessels is isolated from the flowing stream and the contained fluid is depressurized to no less than the vapor pressure of brine at 150°C (4.7 atm for pure water) to permit CH4 exsolution without flashing of the brine. Concurrently, in the other vessel, brine (which has already been depressurized) is expelled to the injection well by displacement with fresh brine under pressure from the wellhead separator. To prevent mixing, a mobile barrier must exist between the spent and fresh fluid volumes.

When these steps are completed in the respective vessels, the vessel function is reversed to maintain continuous operation. Well flow is not interrupted, dissolved CH4 is recovered, and direct injection of the spent brine without pumping is accomplished by proper valve sequencing. The process takes place at near isothermal and adiabatic conditions and does not preclude extraction of the thermal energy, if desired. The Fig. 5 schematic diagram illustrates the basic flow streams and valving required for the sequential depressurization-repressurization process.

There are several aspects of the process that will require study and development. Methane exsolution rates and the dynamics of gas-liquid disengagement will require evaluation in laboratory tests. Mixing of the rich and spent brines must be held to an absolute minimum to ensure high overall CH4 recovery. We are presently considering use of flexible membranes to meet these design requirements. We will also need to develop the valve-sequencing operation and the control system for depressurizing the vessels for CH4 recovery. The most practical shape of the vessels may not necessarily be spherical, but clam-shaped instead to permit higher membrane cycle frequencies in

order to reduce displacement volumes and hence vessel size. It is conceivable that vessel sizes may remain large because of limitations in membrane cycling frequencies or the kinetics of CH₄ exsolution. Being pressure vessels though, it is obviously desirable to minimize their volume.

Hydraulic-Driven Reciprocating Piston Pump

A coupled, dual piston arrangement with a separate low-pressure vessel for CH4 exsolution is shown in Fig. 6. This arrangement presumably overcomes the membrane system difficulties. It separates the CH4 exsolution step from the pumping operation and by virtue of the piston-cylinder design permits higher cycle frequency and lower pump displacement volumes.

Venturi Nozzle

The pressure regimes developed in a venturi nozzle may be advantageously utilized in this particular application. In a continuously flowing stream through the nozzle, the low-pressure regime at the throat would permit CH, exsolution, followed by liquid-pressure recovery on the order of 70% in a properly designed divergent section. Anticipated problems would be flow instabilities produced by continuous CH, exsolution in the converging section of the nozzle, gas separation and removal problems in the throat, and entrance flow instabilities in the divergent section. Scale formation in the nozzles could also be a serious problem.

Hydraulic Turbines

Other possibilities include use of hydraulic turbines or gear drives coupled directly to injection pumps, as shown in Fig. 7. Methane exsolution begins in the turbine chambers and is completed in a separator vessel. Velocity-pump reaction turbines and variations specifically modified for expansion of geopressured fluids are described by Austin and House¹⁸ and may be applicable in driving an injection operation.

All of these devices have mechanical and hydraulic losses that need to be evaluated. The ultimate choice may not be based on the most efficient machine, but rather on the most reliable system capable of performing in the potentially corrosive and scale-forming geopressured-brine environment.

CONCLUSIONS

Based on economic assessments, 1,2 exploitation of the geopressured resource for CH4 recovery does not look promising, unless CH, selling prices rise steeply relative to production costs. A significant but not overriding reduction in costs can be accomplished by utilizing the pressure at the wellhead/gas separator for direct injection into shallow aquifers, thereby eliminating or greatly reducing OaM costs for injection pumping. If it becomes necessary to inject into the production reservoir to maintain productivity and minimize subsidence, maintenance of higher than normal wellhead pressures could be adopted to significantly reduce injection-pump work requirements. This could be cost effective even with reduced production rates. However, the option of pressure maintenance has economic benefit only with recovery of that remaining portion of CH_4 still dissolved at elevated pressures. Extraction of CH_4 at high temperatures and pressures may be accomplishable by solvent extraction techniques.

Gas stripping is another technically feasible method, but does not appear economical because of gas dissolution losses. Mechanical devices also show promise. These methods are variations of positive displacement hydraulic motors and hydraulic turbines with separators or other provisions to permit CH, exsolution and subsequent recovery. All of these techniques would operate at near isothermal conditions, which would permit thermal energy recovery if desired. More detailed analyses of these methods including measurements of mutual solubilities, tests of small extraction columns, design and tests of prototype machines and separators, and estimates of process costs, are now underway.

ACKNOWLEDGMENT

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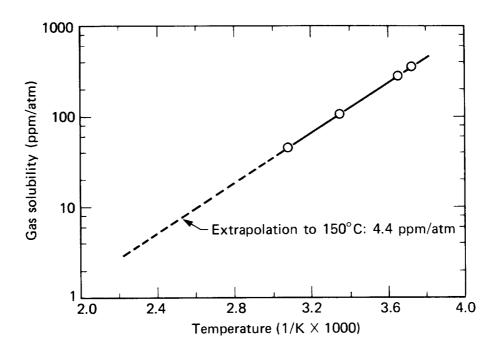
TABLE 1

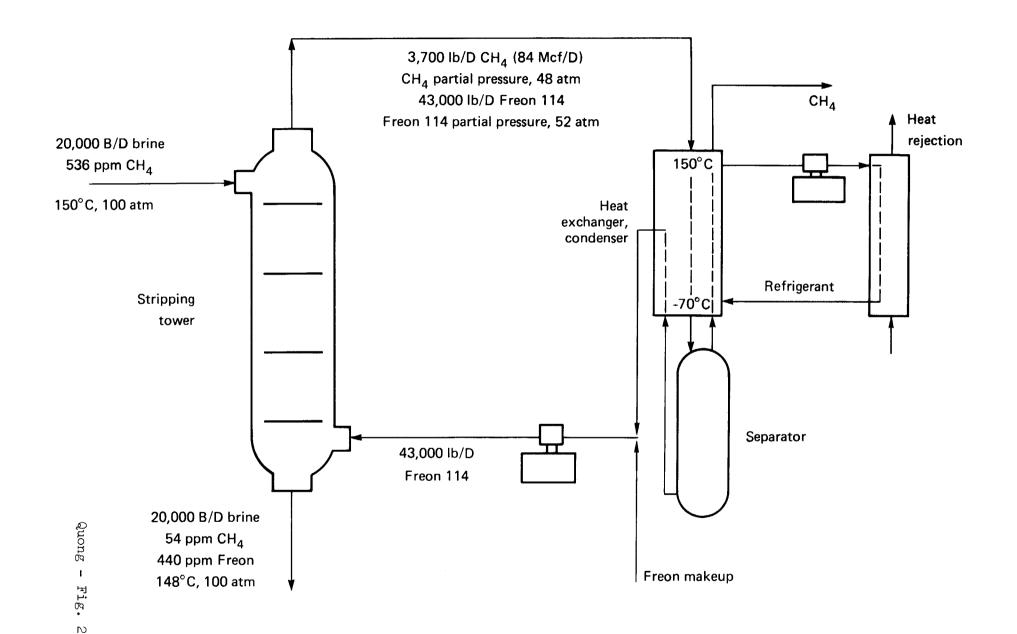
PARAMETERS USED TO ARRIVE AT A PROFITABLE SELLING PRICE OF METHANE

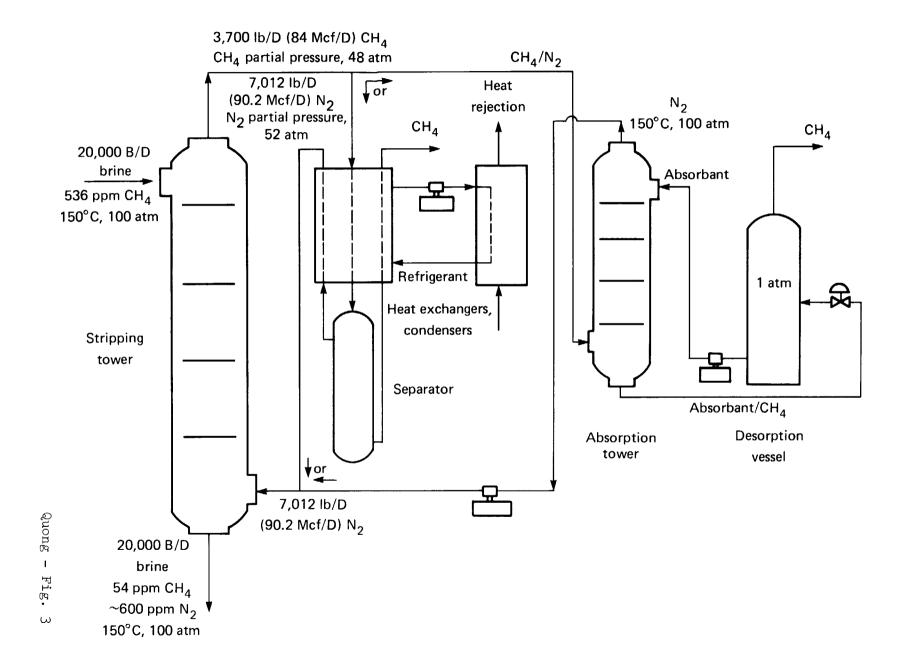
	Stu	ıdy
Parameter	Swanson and Osoba ¹	Doscher et al. ²
Reservoir pressure, psi	10,318	11,000
Drainage area, mi²	12.6	11.0
Permeability, md	15	18
Porosity, %	20	21.6
Compressibility, psi ⁻¹	6×10^{-6}	1.1×10^{-5}
Thickness, ft	230	162
Well diameter, ft	0.458	0.400
Well depth, ft	15,560	13,000
Fluid viscosity, cp	0.20	0.236
CH4/bbl fluid, scf/bbl	40	40
Av flow rate, B/D	9,700	9,956
Well life, years	20	20
Required profitable selling price	\$7.50/Mcf (includes recovery of \$0.05/bbl for thermal energy component)	\$9.24/Mcf (before federal taxes)

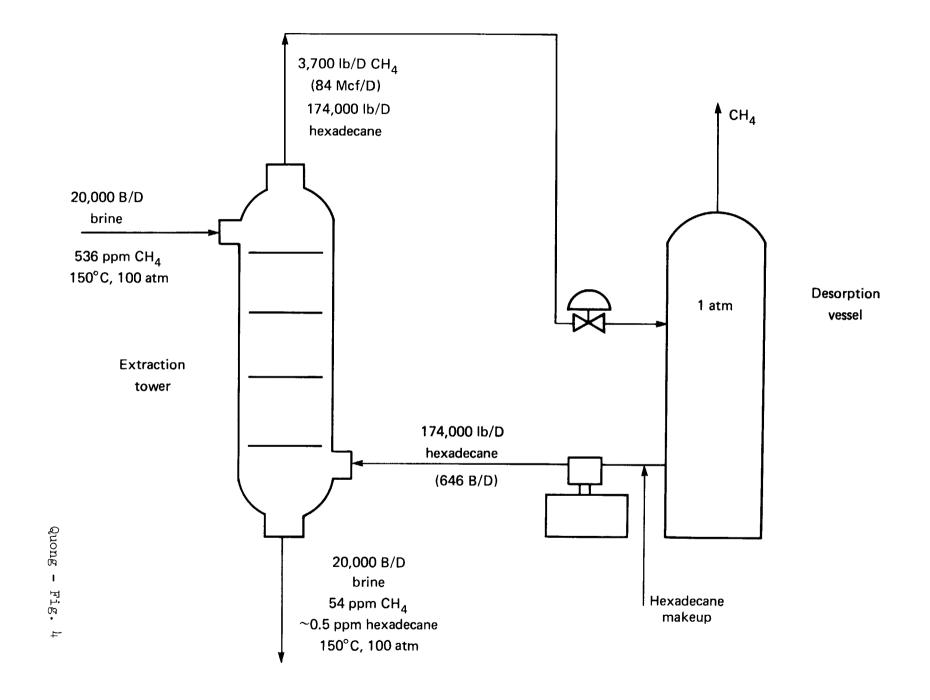
FIGURE CAPTIONS

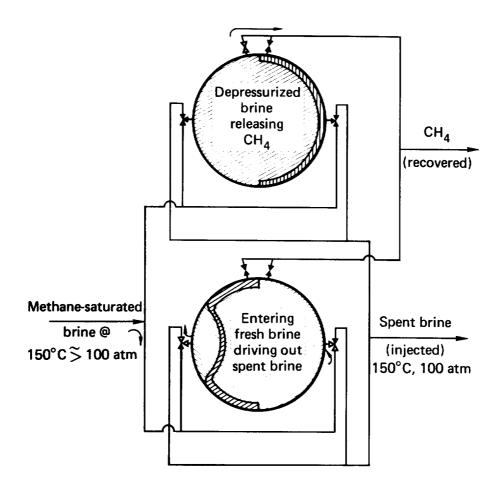
- Fig. 1-Freon 114 gas solubility in NaCl solution.
- Fig. 2-Methane stripping from geopressured brines using Freon 114.
- Fig. 3-Methane stripping from geopressured brines using $\rm N_2$ gas. Purification by liquefaction or absorption.
- Fig. 4-Methane extraction from geopressured brines by liquid extraction.
- Fig. 5-Sequential depressurization-repressurization process for ${\rm CH_4}$ desorption and brine injection in geopressured systems.
- Fig. 6-Hydraulic-driven piston pump with a ${\rm CH_4}$ separator.
- Fig. 7-Coupled hydraulic turbine-pump with a ${\rm CH_4}$ separator.



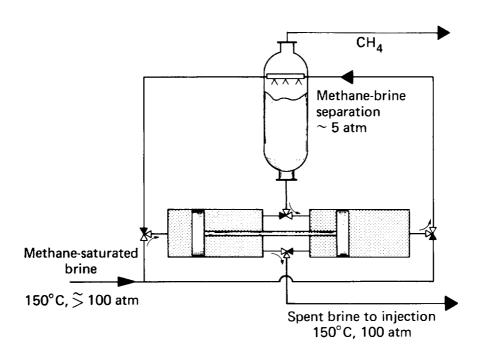








Quong - Fig. 5



Quong - Fig. 6

